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## Oscillatory exchange bias in Fe/Cr double superlattices

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## Abstract

In the  $[Fe/Cr]^{AF}/Cr_x/[Fe/Cr]^F$  double superlattices consisting of a ferromagnetic Fe/Cr superlattice on top of an antiferromagnetic Fe/Cr superlattice, the exchange coupling between the superlattices is determined by the thicknesses (x) of the Cr spacer layer. The oscillating behavior of the exchange bias field of a series of (2 1 1)-oriented Fe/Cr double superlattices was determined by superconducting quantum interference device (SQUID) and magneto-optic Kerr effect (MOKE) measurements. For x > 13 Å a negative strongly oscillating character of the exchange bias was observed. At very thick x the exchange bias vanishes. The most immediate result is the fact that the exchange bias field is always negative, regardless of the sign of the coupling between the ferromagnetic and the antiferromagnetic superlattices. The detailed dependence of the exchange bias field as a function of the intersuperlattice thickness of Cr is explained in terms of the interaction between the two superlattices in collinear configuration. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Exchange bias; Superlattice; Interlayer coupling; Exchange coupling; Epitaxy

Exchange anisotropy is an effect caused by the magnetic interface interaction between a ferromagnet (F) and an antiferromagnet (AF) [1]. The ferromagnet magnetization (M-H) loop shift away from H = 0 indicates the existence of an exchange anisotropy. The magnitude of this shift is know as the exchange bias field  $H_E$ .  $H_E$  depends stronly on many factors, including the spin structure at the interface, the antiferromagnetic anisotropy, roughness, crystallinity, etc. [2]. The detailed dependence from these factors has been the subject of an enormous experimental and theoretical endeavor during the past years. The important technological

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applications of this effect make its study of even greater interest. The exchange bias effect is utilized for permanent magnet materials [3], high-density recording media [4] and domain-stabilized recording heads [5].

In order to better understand the fundamental aspects of exchange bias we have studied the shift of the hysteresis loops in *double superlattices*, which are artificial magnetic systems where the exchange bias can be realized and analyzed with minimal material-related complexities [6]. The working of these novel systems are constructed based on the well-established oscillatory interlayer exchange coupling in Fe/Cr so that the ferromagnetic superlattice on top of the system is coupled ferromagnetically or antiferromagnetically to the antiferromagnetic superlattice multilayer at the

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bottom. The Cr spacer layer thickness between the two superlattices is varied to provide a controlled exchange coupling between the AF and F superlattices.

The double superlattice structure has many advantages over conventional systems for the study of the exchange bias effect. Since the exchange bias effect is primarily an interface phenomenon, the first requirement for a systematic investigation is to have highly ideal AF/F interfaces that are easy to characterize and to manipulate. This is satisfied in the double superlattices: the long period of 18 Å for interlayer coupling in Fe/Cr makes the doublesuperlattice structure less sensitive to roughness and amenable to characterization techniques such as polarized nuetron reflectivity measurement. A second prerequisite for exchange bias is magnetic anisotropy in the AF region. For double-superlattice systems this may be obtained as a growthinduced uniaxial in-plane magnetic anisotropy. Fe/Cr (211) superlattices can be epitaxially sputtered on single-crystal MgO (110) substrates, with the easy axis along the Fe/Cr  $[0\overline{1}1]$  direction [7]. In this paper we present the behavior of the exchange bias field in Fe/Cr(211) double superlattices as a function of the thickness x of the Cr spacer layer between the two superlattices, and relate it to the exchange coupling between the two superlattices.

At first thought the exchange field  $H_{\rm E}$  is strictly proportional to  $J_{\rm int}$ , the exchange energy between the two superlattices. In Fig. 1 is shown that, at least for a magnetic configuration where AF and F magnetization vectors are collinear,  $H_{\rm E} \propto -|J_{\rm int}|$ . This is why in almost all cases, in which the exchange bias has been experimentally observed in AF/F pairs, this turned out to be negative [1,3,5]. However,  $H_{\rm E} > 0$  in certain FeF<sub>2</sub>/Fe [8] and  $MnF_2/Fe$  [9] bilayers when the systems are cooled through the Néel temperature of the AF in a large positive field. Positive exchange bias has been attributed to competition between the antiferromagnetic coupling at the AF/F interface and the Zeeman energy of the AF surface spins [9]. Up to now it was assumed that the coupling  $J_{int}$  is the weak link, being smaller than both the interlayer coupling in the F superlattice,  $J_{\rm F}$ , as well as the similar quantity for the AF superlattice,  $J_{AF}$ . Suppose that  $J_{int}$  is considerably stronger than  $J_F$ . Then, upon switching of the field, the magnetic configuration that is energetically most favorable is one in which a domain wall (or partial domain wall) is created in the ferromagnet or the antiferromagnet [10-12]. Also, these hypothetical configurations have to be compared with a scenario, in which switching takes place with the creation of lateral domains [13]. With the double-superlattice structure, we can simply vary the Cr spacer thickness to vary  $J_{int}$ , and to explore the interplay between  $J_{int}$ ,  $J_{\rm F}, J_{\rm AF}$  and the Zeeman energy.

The double superlattices were grown via DC magnetron sputtering onto single-crystal MgO (110) substrates. The samples had a layer sequence  $Cr_{50 \text{ Å}}/[Cr_{20 \text{ Å}}/Fe_{50 \text{ Å}}]_5/Cr_x/[Fe_{14 \text{ Å}}/Cr_{11 \text{ Å}}]_{20}/Cr_{200 \text{ Å}}/MgO(110)$ . The 200 Å Cr buffer layer deposited at 400°C established good epitaxy with the

$J_{Int} > 0$		<b>J</b> <sub>Int</sub> < 0	
$H > +\epsilon$	$H < \textbf{-} J_{int} /N_F t_F M_F$	$H > +\epsilon$	$H < \textbf{-} J_{int} /N_F t_F M_F$
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	<b>4</b>	>	4
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		<b>4</b>	4
4	4	<b>&gt;</b>	
	·····	<b>4</b>	<b>4</b>
<b>4</b>	<b>4</b>		

Fig. 1. Sketch representing the spin configurations of a double superlattice before  $(H > +\varepsilon, \varepsilon)$  is a small but finite value) and after  $(H < -|J_{int}|/N_F t_F M_F)$  switching. The left panel is for ferromagnetic inter-superlattice coupling  $(J_{int} > 0)$  and the right panel is for antiferromagnetic inter-superlattice coupling  $(J_{int} < 0)$ . In both cases, the exchange bias field is negative because the top Fe layer in the AF superlattice adopts a direction compatible with the sign of  $J_{int}$ .

substrate. The double superlattice structure was grown at 90°C and covered on top by a 50Å Cr laver. The thickness of the Cr spacer laver between the F and AF superlattices, x, was varied from 11 to 70 Å to cover four oscillations of the sign of the inter-superlattice coupling. Care was taken to ensure that the samples within the series are otherwise identical. Structural characterization of all prepared samples was done by small and high angle X-ray diffraction using  $Cu K_{\alpha}$  radiation. Even at large Cr spacer layer thicknesses the low-angle Xray diffraction peaks were clearly resolved, indicating that the films are well-layered. The Bragg peaks have a FWHM of 0.37-0.58° which leads to coherence lengths of 150-234 Å. The typical values for layer roughness are shown in Ref. [14].

The magnetization measurements of all prepared samples were obtained with a superconducting quantum interference device (SQUID) magnetometer and a magneto-optic Kerr effect (MOKE) system. The measurements were taken at room temperature and with the magnetic field applied along the easy axis. With the SQUID magnetometer we measured both full and minor hysteresis loops. The bias effect in the double superlattice is obtained by aligning the magnetization of both F and AF superlattices in a high field of 15 kOe and then run the minor hysteresis loop. The shifted hysteresis loop is the signature of the unidirectional anisotropy. Figs. 2(b) and (c) provide an example of the minor loops of two double superlattices with (a) x = 16 Å,  $H_{\text{E}} = -12 \text{ Oe}$ , and (b) x = 20 A,  $H_{\rm E} = -38$  Oe. In all cases, the minor loops were characterized by very square hysteresis loops of relatively modest width. The value of the exchange bias field  $H_{\rm E}$  was obtained by taking the value of the magnetic field at mid-point of the loop. Fig. 3 gives the dependence of the exchange bias field on the Cr spacer layer thicknesses x. One can see a strong oscillatory character for thin Cr spacer layer thicknesses, consisting of minima and maxima, and the final damping of this oscillations at large Cr thicknesses. The solid line is guide for the eyes. The circles are representing the ferromagnetically coupled interfaces and the squares are representing the antiferromagnetically coupled interfaces.

On the basis of detailed neutron reflectivity measurements [6,14] on one of the sample it was

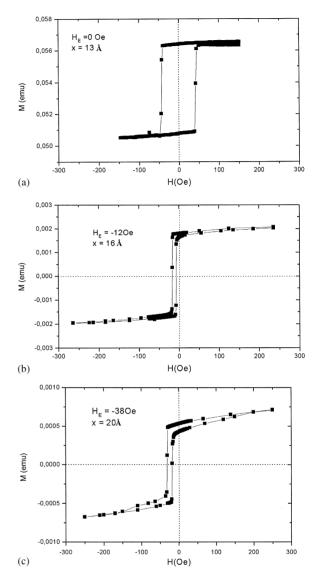


Fig. 2. Magnetization minor loops for double superlattices with: (a) x = 13 Å and  $H_E = 0$  Oe; (b) x = 16 Å,  $H_E = -12$  Oe; (c) x = 20 Å,  $H_E = -38$  Oe.

concluded that the antiferromagnetic structure was collinear to the ferromagnetic structure. It was also confirmed that in the two states of the minor loop the AF superlattice was fixed and only the spins of the F superlattice were switched. The magnitude of the exchange bias field was found to be equal to the value expected of the exchange interaction between collinear AF and F superlattices [6]. Thus, it was

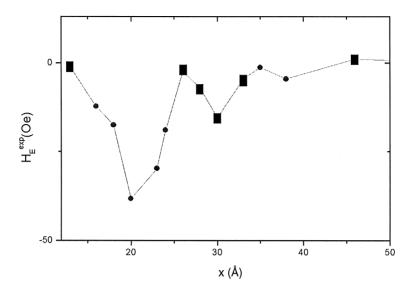


Fig. 3. The exchange bias field  $H_{\rm E}$  as a function of the Cr spacer layer thicknesses, x, for the analyzed double superlattices. The solid line is guide to the eyes. The circles are representing the F-coupled interfaces and the squares are representing the AF-coupled interfaces.

justified to use the classic formula for the magnitude of the exchange bias field as applied to systems of collinear spin structures  $H_{\rm E} = -|J_{\rm int}|/N_{\rm F}t_{\rm F}M_{\rm F}$ , where  $N_{\rm F}$  is the number of Fe layers in the ferromagnetic superlattice, and  $t_{\rm F}$  and  $M_{\rm F}$  are the thickness and saturation magnetization of the Fe layers. The obtained oscillatory behavior of the exchange bias field versus the Cr spacer layer thicknesses follows the oscillatory interlayer coupling in Fe/Cr (211) superlattices presented in Ref. [7]. However, since the double superlattices do not require cooling in a field to establish exchange bias, the top Fe layer in the AF superlattice adopts a direction compatible with the inter-superlattice coupling as illustrated in Fig. 1. Therefore,  $H_{\rm E}$  is always negative regardless of whether the inter-superlattice coupling is ferromagnetic or antiferromagnetic.

At thin Cr spacer layer thicknesses (x = 13 A) the experimental value of  $H_E$  is zero within experimental error. However, the MOKE hysteresis loop represented in Fig. 2(a) is very wide, with coercivity  $H_c = 42$  Oe. The value of  $H_c$  is comparable to the calculated value for  $H_E$  using the classical formula for collinear spin structures mentioned above ( $H_E = 94.1$  Oe). This suggests that the AF superlattice has switched irreversibly during the minor loop measurement [15]. The enhancement of coercivity in conventional exchange biased systems has been associated with instability of antiferromagnetic grains in recent theories [16,17]. Our observation in double superlattices with very thin Cr spacers is consistent with them.

In conclusion, we report the observation of an oscillatory behavior of the exchange bias field depending on the Cr spacer layer thicknesses between an antiferromagnetic and a ferromagnetic superlattice in novel double superlattice structures. The oscillatory behavior is similar to the oscillatory dependence of the coupling strength in Fe/Cr (211)superlattices, which is a clear indication of a linear dependence of the exchange bias field on the interfacial exchange energy as it was expected. A striking effect is the observation of exchange bias fields which is independent of the sign of the coupling. With double superlattices the AF/F coupling can be controlled by varying the spacer thickness as demonstrated. This ability to fine tune the bias field could have implications on applications.

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